

Dirty Superconductivity in the Electron-Doped Cuprate $\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_{4-\delta}$: a Tunneling Study

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(Dated: February 6, 2008)*

We report a tunneling study between $\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_{4-\delta}$ and Lead as a function of doping, temperature and magnetic field. The temperature dependence of the gap follows the BCS prediction. Our data fits a nonmonotonic d -wave order parameter for the whole doping range studied. From our data we are able to conclude that the electron-doped cuprate $\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_{4-\delta}$ is a weak coupling, BCS, dirty superconductor.

PACS numbers: 74.50.+r 74.20.Rp 74.62.Dh

In the theory for superconductivity by Bardeen, Cooper and Shrieffer (BCS) as the temperature is raised from $T=0$ K the amplitude of the superconducting order-parameter decreases and eventually becomes zero at the critical temperature T_c [1]. This is not necessarily the case for the high T_c cuprate superconductors. For example scanning tunneling spectroscopy showed a non vanishing gap above T_c and a constant gap amplitude below T_c for hole doped cuprates [2]. It has been proposed [3] that in the high T_c cuprates the amplitude of the order-parameter does not go to zero at T_c but phase fluctuations eventually destroy long range coherence. The Uemura plot [4], showing that T_c scales with the superfluid density gave further support to this scenario. The electron-doped cuprates, however, fall off the Uemura line [5] and this raises the fundamental question whether the order-parameter falls to zero at T_c for these compounds.

The symmetry of the order-parameter in the electron-doped cuprates is still a matter of debate. While many experiments suggest an order-parameter having a d -wave symmetry [6, 7, 8, 9] others suggest a change of symmetry with doping [10, 11]. Raman spectroscopy on optimally doped samples has been interpreted in terms of non-monotonic d -wave ($\text{nm}d$) in which higher harmonics have significant contribution [12]. In this case the amplitude of the order-parameter has a maximum at angle smaller than 45° to the nodal direction. Further support for this scenario was later found experimentally from angle resolved photoemission spectroscopy [13] and from theoretical calculations [14]. Yet, it is still a mystery why the tunneling spectra for electron-doped cuprates are isotropic in the ab plane [15] and missing the expected zero bias conductance peak in low transparency junctions [16]. Furthermore, for such order-parameter any scattering center is a pair breaker that destroys superconductivity in its vicinity [17]. For hole doped cuprates the coherence length, ξ_{BCS} is usually much shorter than the mean free path ℓ , thus allowing the order-parameter to recover between scattering events. For electron-doped cuprates ξ_{BCS} is an order of magnitude larger than for

the hole doped. It is therefore important to find out whether these compounds are in the clean ($\xi_{BCS} \ll \ell$) or dirty regime.

In this letter we report a tunneling study as a function of doping, temperature, and magnetic field using lead/Insulator/ $\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_{4-\delta}$ (Pb/I/PCCO) planar junctions. We find that the order-parameter goes to zero in a BCS like fashion. This rules out phase fluctuations as the reason for loss of coherence at T_c . We obtain a good fit to a non-monotonic d -wave order-parameter in the PCCO electrode for the whole doping (up to $x=0.19$) and temperature range studied. From our tunneling data we are able to conclude that PCCO is in the dirty limit. This suggests a possible solution for the long standing puzzles of tunneling isotropy and the absence of zero bias conductance peak in electron-doped cuprates.

In a tunneling experiment a quasiparticle is injected into a superconductor through a dielectric barrier. This method has been proven to be a powerful tool for probing the density of states in conventional superconductors [18]. A theory was later developed to account for barriers of various transparencies using a single parameter Z [19]. It was later extended for the case of anisotropic materials and order-parameters [16].

Lead counter electrodes were deposited on PCCO c -axis oriented films using a method described elsewhere [20]. This procedure results in good tunnel junctions with a lead-oxide barrier [21]. At zero field and below $T_c(\text{Pb})$, we obtain superconductor/I/superconductor (SIS) tunneling spectra. These spectra exhibit conductance close to zero at low biases and Pb phonon signatures, indicating good tunnel junctions [18]. This is different than grain boundary junctions where the leakage current was reported to depend on the doping level [22]. Previously, we showed that the spectra obtained on ab faces of single crystals are identical to those of c -axis oriented films. This suggests dominating in plane tunneling for the latter case [20]. The presence of in-plane nano-facets and a much reduced tunneling probability in the c direction [23] can be the reason for the in-plane dominance. Upon

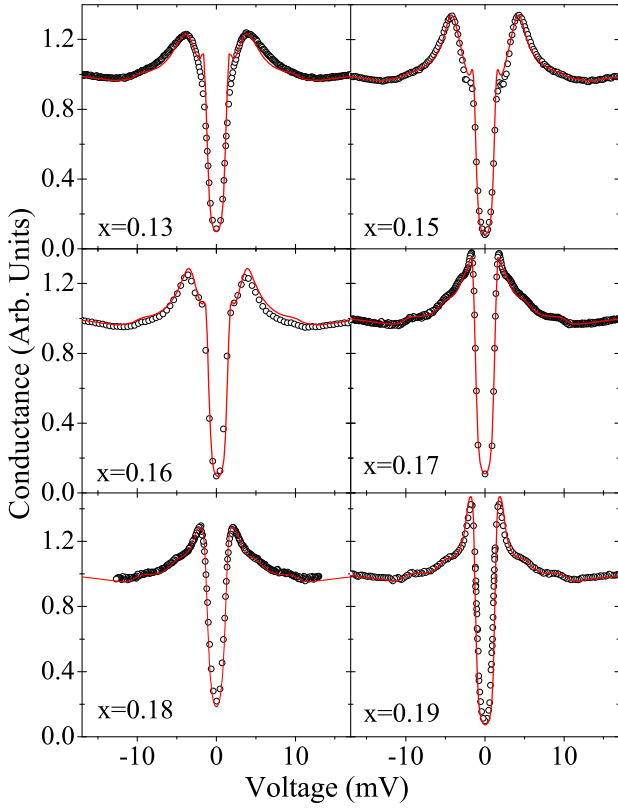


FIG. 1: (color online) Conductance versus voltage for the six doping levels of $\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_{4-\delta}$ studied at 1.8K and zero field (circles). At this temperature and field both electrodes are superconducting; note the Pb phonons around 5 and 9 meV. The red solid line is a non-monotonic d -wave fit to the data described in the text.

applying a 14T magnetic field perpendicular to the ab planes and to the junction, one drives both electrodes into their normal (N) state. This allows us to normalize each spectrum with its respective 14T one, thus cancelling extrinsic effects coming from either the junction or the counter electrode. This is the standard procedure used in classical tunneling experiments[18]. This procedure was repeated for magnetic fields up to 14T and temperatures up to 30K. We define $T_c(\text{PCCO})$ at the junction as the temperature at which the zero field spectrum merges with its corresponding 14T one. The upper critical field, H_{c2} , at 1.8K is the field at which the spectrum merges with that of the 14T one.

In Fig.1 we show the normalized S/I/S spectra (circles) at $T=1.8\text{K}$ and $H=0\text{T}$ for doping levels ranging from underdoping ($x=0.13$) through optimum doping ($x=0.15$) to heavy overdoping ($x=0.19$). To fit the data a tunneling conductance, $G(eV)$, can be calculated as follows:

$$\frac{G(eV)}{G_n} = \frac{d}{d(V)} \int D_1(E, T) D_2(E + eV, T) \times [f(E, T) - f(E + eV, T)] dE \quad (1)$$

where, G_n is the conductance when both electrodes are in the normal states, V is the bias across the junction, f is the Fermi distribution function at a given temperature, T . D_1 is the tunneling density of states calculated as in Ref.[19] for the well known Pb density of states including the phonon spectrum. D_2 is tunneling density of states calculated for PCCO using the theory for anisotropic order-parameter in Ref.[16] after integrating over all angles using the fitting parameters: Z , Δ_0 , η , and Γ , where Z is the barrier strength, Γ is a life-time broadening[24]. We introduce the following form for a nmd order-parameter:

$$\Delta(\theta) = \Delta_0 \cos(2\theta) \left\{ 1 + \frac{\eta \cos^2(2\theta)}{1 - 0.9|\cos(2\theta)|} \right\}^{-\frac{3}{2}} \quad (2)$$

where θ is the angle between quasi-particle momentum and the (1,0,0) direction. This function keeps the nodes and phases as in the case of pure d -wave but shifts the gap maximum towards the node with increasing η . The solid line in Fig. 1 is the best fit obtained with the parameters described in table I. The gap amplitude Δ_{max} and the angle from the 100 direction at which the maximum gap is obtained θ_{max} are in agreement with photoemission measurements[13]. Such an order-parameter fits the data better than a pure d -wave, much better than s -wave or any other linear combination of the two.

For $x=0.13$ and $x=0.15$ we fit the tunneling characteristics up to T_c . We use a BCS temperature dependence for the Pb order-parameter and the measured temperature T . We keep all other parameters used for 1.8K, constant varying only a single parameter Δ_0 . In Fig.2 we show examples for such fits below and above $T_c(\text{Pb})=7.2\text{K}$. The remarkable agreement between the data and the fits give further support to the fitting parameters used at the base temperature since they fit both the S/I/S and the N/I/S cases. For the latter the calculation simply reduced to that of Ref.[16]. We follow the temperature dependence of Δ_0 obtained from the fits. This allows us to find $\Delta_{max}(T)$ from Eq. 2. The obtained $\Delta_{max}(T)$ for $x=0.13$ (squares), $x=0.15$ (circles) are plotted on the universal BCS graph in Fig.3. We note a very good agreement to the BCS prediction (dashed line)[1].

The agreement with the BCS temperature dependence is in contrast with the hole doped cuprate data [2]. This behavior rules out phase fluctuations as the cause for the disappearance of long range order superconductivity at T_c . This is consistent with the absence of a strong Nernst signal well above T_c [25] and the absence of an extended pseudogap phase [20] in electron-doped cuprates.

Next we calculate the BCS parameters inferred from our data (see Table I). First, we find $2\Delta_{max}/kT_c$ for the various doping levels, where the low temperature values of Δ_{max} are used. Within experimental errors, our results yield the BCS weak-coupling ratio of 3.5. Unfortunately, there is no available theoretical estimation of $2\Delta_{max}/kT_c$ for the nmd case. Second, we cal-

TABLE I: Superconducting parameters for the various junctions. The critical temperature T_c and field H_{c2} are measured using the tunneling conductance as described in the text. All other parameters are found at 1.8K. Δ_{max} is the gap amplitude reached at an angle θ_{max} measured from the a-axis. $\Delta_{max}, \theta_{max}$ are found using the fitting function, described in the text. For a simple d -wave order-parameter $\theta_{max} = 0$. ξ_{GL} and ξ_{BCS} are the Ginzburg-Landau and the BCS coherence lengths respectively. ℓ can be calculated from the latter two length scales.

Ce doping	$T_c(K)$	$\mu_0 H_{c2}(Tesla)$	$\Delta_{max}(meV)$	$\theta_{max}(deg)$	Z	$\Gamma(meV)$	$\frac{2\Delta_{max}}{kT_c}$	$\xi_{GL}(nm)$	$\xi_{BCS}(nm)$	ℓ (nm)
0.13	17 \pm 1	11 \pm 1	2.5	16	4	1.3	3.5 \pm 0.5	5.5 \pm 0.3	31 \pm 9	1.3 \pm 0.4
0.15	19 \pm 1	7 \pm 1	3.25	18	4	0.9	4.0 \pm 0.4	6.9 \pm 0.5	24 \pm 6	2.7 \pm 0.8
0.16	16 \pm 1	5 \pm 1	2.6	18	4	0.95	3.8 \pm 0.5	8.1 \pm 0.8	30 \pm 8	3 \pm 1
0.17	13 \pm 1	3 \pm 1	1.3	22	4.1	0.93	2.3 \pm 1.3	10.5 \pm 1.7	61 \pm 21	2.5 \pm 1.2
0.18	11 \pm 1	1 \pm 0.2	1.0	22	7	0.75 ^a	2.1 \pm 1.5	18 \pm 2	79 \pm 31	5.7 \pm 2.5
0.19	8 \pm 0.4	0.3 \pm 0.1	0.9	23	4.9	0.45	2.6 \pm 0.9	33 \pm 6	87 \pm 36	17 \pm 9

^aAn additional life-time broadening of 0.25meV was added to the Pb counter electrode.

culate $\xi_{BCS} = \frac{\hbar v_F}{\pi \Delta_{max}}$ where the averaged Fermi velocity v_F can be estimated from photoemission measurements to be $3.75 \cdot 10^5$ m/sec.[26]. Using H_{c2} evaluated earlier, we calculate the Ginzburg-Landau coherence length, $\xi_{GL} = \sqrt{\frac{\phi_0}{2\pi H_{c2}}}$, with ϕ_0 the flux quantum. Our results are in agreement with Raman spectroscopy data [27].

Despite the fact that hole-doped cuprates are considered to be “bad metals” in their normal state, they are still in the clean limit. This is due to their short coherence length [28]. Here, we find a BCS coherence length an order of magnitude larger than in the hole-doped case. Therefore, it is possible that electron-doped cuprates are closer to the dirty limit, *i.e.* $\xi_{BCS} > \ell$, where ℓ is the mean free path. Since ξ_{GL}/ξ_{BCS} is well below its clean limit value of 0.74, we roughly estimate (see Table I) ℓ using the relation for the dirty limit $\xi_{GL} = 0.855(\xi_{BCS}\ell)^{1/2}$ [29]. Indeed, we find that we are in the dirty limit. Recently, Homes *et.al.* came to similar conclusions from optics measurements on $\text{Pr}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-\delta}$ samples [30].

We can independently estimate ℓ from resistivity using $k_F \ell = \frac{\hbar d \sigma_{ab}}{e^2}$ to verify our results. Here, d is the distance between two successive CuO_2 planes, σ_{ab} is the in-plane conductivity, and e is the electron charge. First, we notice that the doping dependence of ℓ obtained from tunneling (within the error bars) is consistent with that of the resistivity [31]. Second, we can obtain an upper limit for ℓ by estimating the Fermi wave number, $k_F \simeq \frac{mv_F}{\hbar}$ using m as the bare electron mass and v_F from photoemission measurements [26]. For $x=0.19$ we obtain from the resistivity at 0.3 K, $\ell = 23\text{nm}$, in excellent agreement with the tunneling estimation.

Our tunneling spectra are very different from those obtained on hole-doped $\text{YBa}_2\text{Cu}_3\text{O}_7$ (YBCO) using the same counter electrode (Pb) [32]. First, while the data in Fig 1 exhibits conductance peaks at $\Delta(\text{PCCO}) \pm \Delta(\text{Pb})$ as in classical tunneling experiments [18], for the case of in-plane tunneling into YBCO the peaks appear at $\Delta(\text{YBCO})$ and $\Delta(\text{Pb})$ separately. Moreover, strong in-

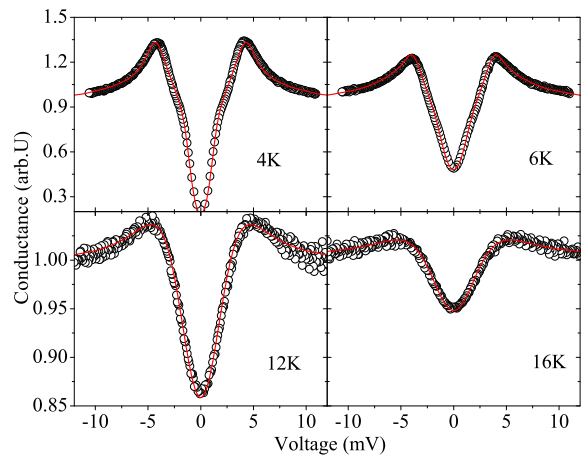


FIG. 2: (color online) Lead / $\text{Pr}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-\delta}$ tunneling spectra at representative temperatures (circles). The line is a fit using the parameters obtained from Fig 1. with the gap amplitude ($\Delta_0(T)$) being the only fitting parameter.

plane anisotropy is observed for various film orientations in YBCO in contrast to PCCO [15].

Although our experiment alone can not exclude a possible anisotropic s -wave order-parameter, the tunneling fits are consistent with phase sensitive experiments for optimum doping [6] and slight over-doping [9] that indicate the existence of nodes and phase changes in the order-parameter. However, d -wave or nmd order-parameters should result in a zero-bias conductance peak for in-plane tunneling spectroscopy. This zero-bias conductance peak has never been observed in low transparency junctions ($Z > 1$) in contrast with hole-doped cuprates where it can be hardly avoided [33, 34]. Another puzzle is lack of directional in-plane sensitivity in tunneling measurements [15]. These discrepancies between quasiparticle tunneling [15] and Cooper pair tunneling

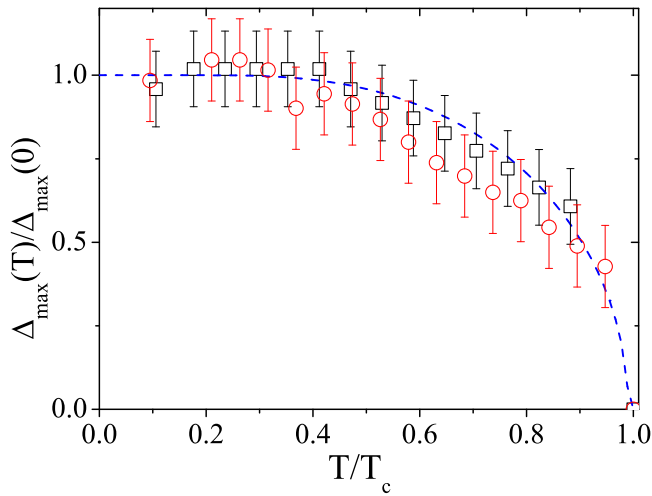


FIG. 3: (color online) The reduced gap maximum obtained from fits as demonstrated in Fig.2 plotted *versus* reduced temperature for $\text{Pr}_{1.87}\text{Ce}_{0.13}\text{CuO}_{4-\delta}$ (squares) and $\text{Pr}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-\delta}$ (circles). The dashed line is the BCS universal line.

[6, 9] may be resolved assuming dirty superconductivity as we report. For the first case, quasiparticles undergo several scattering events within a coherence length and thus become insensitive to their initial momentum and phase. Therefore, the initial direction from which the quasiparticles are injected is unimportant. By contrast, Cooper-pairs retain their phase information over a coherence length scale and consequently experiments involving their tunneling can probe the anisotropic phase of the order-parameter. However, it still deserves theoretical attention to resolve the question how an order-parameter which changes sign, such as nmd , can still survive in the dirty limit. For such an order-parameter any scattering center is a pair breaker [17].

In summary, we measured tunneling conductance for $\text{Pb}/\text{Insulator}/\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_{4-\delta}$ over wide doping, temperature and magnetic field ranges. Taking advantage of the accessible upper critical field of the electron-doped cuprates, the well studied superconducting parameters of lead, and the clear features in S/I/S contacts, we were able to properly normalize and fit the data even at relatively high temperatures. The data fits a non-monotonic d -wave behavior (for PCCO) over the entire doping range. The gap maximum follows the BCS temperature dependence for underdoped and optimally doped samples. The upper critical field extracted from our tunneling data and the gap amplitude are used to calculate the BCS and the Ginzburg-Landau coherence lengths. From these length scales we estimate the mean free path and conclude that PCCO is in the dirty limit. This may explain the absence of a zero bias conductance peak in low transparency junctions and the lack of in-plane directional sensitivity in the tunneling spectrum.

We thank G. Deutscher and G. Blumberg for useful discussions and A. Chubukov for many discussions and for suggesting the nmd fitting function. Support from NSF grant number DMR 0352735 is acknowledged. R. B. acknowledges support from Israeli Science Foundation and the Heinrich-Hertz Minerva Center for High Temperature Superconductivity. Y. D. acknowledges support from the German-Israeli Foundation.

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